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Densified biocoal from woodchips: Is it better to do torrefaction before or after densification?

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HIGHLIGHTS

- Process pathways for making densified biocoal from wood chips were compared in terms of detailed energy and mass balance.
- Binding agent was necessary to densify the torrefied biomass with reasonable energy consumption (process scheme I).
- Raw biomass can be densified first without using any binding agents and can be subsequently torrefied (process scheme II).
- Process scheme II was more efficient in terms of energy/mass balance and resulting pellets had several promising properties.
- Torrefied pellets seems suitable for both biochemical and thermochemical conversion applications.

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ABSTRACT

Torrefied biomass represents a high quality renewable energy commodity that can be used to substitute fossil fuels such as coal. However, densification processes such as pelletisation is necessary to improve the tradability of “low-dense” torrefied biomass. In this work, two process pathways were assessed for energy and mass balance in making torrefied pellets from softwood chips and qualities of the resulting torrefied pellets were compared. Pathway I involve drying the wood chips, torrefaction, grinding followed by densification. In pathway II, wood chips were dried, ground, densified and finally torrefied. The results showed that it was difficult to bind the torrefied biomass particles and a binding agent was necessary to enable their effective pelletisation with reasonable energy consumption. In contrary, pelletization of raw materials was possible without using binding agents and when the “raw wood pellets” were torrefied, the pellets surprisingly stayed intact and had several promising properties such as higher energy/carbon value, reduced moisture content and higher stability in water. In addition, the pathway II was more efficient in terms of overall energy and material balance.

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1. Introduction

Torrefaction is a thermal pretreatment process to upgrade the properties of lignocellulosic biomass to a high quality “energy and carbon carrier” which can be effectively used to substitute fossil fuels [1–4]. The process is generally considered as a mild pyrolysis or carbonisation where the biomass is subjected to temperatures ranging from 200 to 300 °C in the absence of oxygen [5]. The high heating value of the currently marketed wood pellets is approximately 19 GJ per metric ton which is substantially lower that of coal (28–30 GJ per metric ton). This lower heat content limits the proportion of biomass that can be used in co-firing with coal [6,7]. By removing the volatile substances that have a lower heating value from biomass, torrefaction process increases the heat value of the biomass closer to that of coal [8,9]. Shang et al. [10,11] have recently shown that torrefaction increased the higher heating value of Scots pine from 18.4 MJ/kg to 24.3 MJ/kg. In addition to be an effective substitute to coal, torrefied biomass can be processed to both gaseous/liquid fuels and a range of chemicals/materials that are currently derived from petroleum [12–15]. The syngas produced from the gasification of torrefied biomass was found to have a significantly higher quality compared to the syngas

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generated from the raw biomass [16,17]. In addition, torrefying biomass prior to pyrolysis improved the properties of biooil [15,16,18]. Some other beneficial properties of torrefied biomass include their high hydrophobicity and enhanced grindability compared to untreated biomass [19–21]. Hydrophobicity of wood pellets is generally characterised by measuring the moisture uptake in a humidity chamber as well as immersing the pellets in water and determining the degree of water absorption [19–21]. Grindability of pellets is determined by measuring the energy consumption for grinding and the resulting particle size distribution [10,11]. Torrefaction has also been recognised as a robust strategy to overcome the heterogeneity among different types of cellulosic feedstocks thus producing a uniform-quality energy commodity with improved fuel/carbon values [2,22,23].

Despite many advantages, there are several challenges that need to be addressed for the commercial use of torrefied material as a "tradable energy commodity". Bulk density of the torrefied material is often poor; i.e., even lower than that of the raw biomass making the transport and storage of the material economically challenging [24,11,25]. In order to facilitate the effective use of torrefied biomass as a "globally traded bioenergy commodity", the material will have to be densified. It should be noted that it was the recent developments in densification technologies such as pelletisation, that enabled the global transport of biomass as an energy commodity [26]. As a consequence, Canadian wood pellet industry has rapidly evolved as an inevitable part of the global bioeconomy with British Columbia being one of the leading exporters of wood pellets. Therefore, it is apparent that combining the torrefaction and densification processes to make "torrefied wood pellets" has a great potential to upgrade the properties of biomass to a transportable high value energy commodity with several end use applications [8,9,27]. However, torrefied pellets should be produced with minimum energy consumption without compromising the energy and carbon values of torrefied materials while also ensuring that the resulting pellets are stable and durable under varying environmental conditions such as different temperatures and humidity.

There has been some previous work, which has looked into densifying the torrefied biomass. As a result, there are some pilot scale industrial units that produce torrefied wood pellets. Most of this units first torrefy biomass and subsequently densify them. However, the main challenge associated with this traditional approach is the difficulty in densifying biomass particles after they have been torrefied. It was found that torrefied biomass particles were not easy to bind as effectively as the untreated biomass particles [28] and the resulting pellets had poor storage and transport characteristics albeit a greater amount of energy required for pelletisation [20,24,11,28,29,10,30]. An alternative approach used in this work was to torrefy biomass after it has been densified. We have expected that this approach may be more promising compared to the traditional method of making torrefied pellets due to the easiness in integrating a torrefaction as a final step in the existing pellet plants. However, it is not investigated in detail whether wood pellets can retain all of the desirable characteristics after they have been torrefied. Some of the preliminary work has shown that torrefaction can cause a decrease in the durability characteristics of the wood pellets such as compression strength [10,11]. In this work, we have quantified almost all of the relevant parameters of torrefied pellets including density, durability, hydrophobicity, water absorption, fuel value and chemical as well as elemental composition. In addition to understanding the properties of torrefied pellets, it is important to understand the overall mass and energy balance of this proposed process configuration compared to the traditional process configuration, where wood chips are first torrefied and subsequently densified.

The overall objective of the described work was to produce "high quality and tradable biocoal" from biomass by combining torrefaction and densification processes. As was mentioned earlier, detailed comparison of the two process schemes were carried out in the work. The first process scheme followed the traditional method of making torrefied wood pellets which involve drying, torrefying, grinding and pelletisation. The second process scheme involved drying, grinding, pelletisation and subsequently the torrefaction of wood pellets. In this work, a complete mass and energy balance of the two process schemes were carried out together with the detailed evaluation of the resulting wood pellet properties. The work is expected to enable us to choose a better process strategy for producing torrefied wood pellets.

2. Materials and methods

2.1. Raw material

Douglas fir (*Pseudotsuga menziesii*) wood chips with average size of 1.5–2 in. with initial moisture content of 40% (on a wet weight basis) were used as raw material. The sample was collected (Fibreco Company, Vancouver) from a large pile, stored outdoor. Immediately after collection, the samples were sealed in plastic bags and subsequently stored in the cold room (4 °C) to prevent any loss of moisture, minimise biodegradation and off gassing.

2.2. Drying

As received sample, due to its high moisture content was not suitable for direct grinding. Therefore, material was first dried to reach 15% moisture content using the hot air convection oven (Cascade, TEK TFO-28). At first, 3 kg of the wet wood chips were loaded in a tray at a uniform height and subsequently placed in the oven at 110 °C. The duration required to dry the wood chips to 15% final moisture content was determined experimentally by using online TGA measurement. In order to ensure uniform drying, we had only used one layer of wood chips in the trays so there are no top or bottom layers. Subsequently, the total amount of energy required for drying was calculated as the sum of (a) the energy required to heat up both wood chips (H_{wood}), water (H_{water}) and air (H_{air}) to 110 °C and (b) the energy required to evaporate both the free water and a fraction of the bound water till the material reached 15% moisture content (H_{evap}) and finally (c) the heat loss of the drying unit (H_{loss}).

The energy for heating up wood, water and air to 110 °C was calculated by using the equation $MC(T_2 - T_1)$ where M is the dry weight of wood chips/weight of water/weight of air and, T_1 and T_2 are the initial and final temperatures respectively. The specific heat capacity (C) of wood, water and air used in the calculation were 0.28, 4.217, 0.266 kcal per kg C respectively [31].

The amount of energy required to evaporate both the bound and free water till the targeted moisture was calculated by $H_{\text{Evap}} = \text{Water}_{\text{free}}(q_{\text{latent}}) + \text{Water}_{\text{bound}}(q_{\text{latent}} + q_{\text{bound}})$ where the $\text{Water}_{\text{free}}$ and $\text{Water}_{\text{bound}}$ are the respective amount of water to be evaporated to obtain 15% final moisture content and q_{latent} is the latent heat of vapourisation of water and q_{bound} is the additional separation heat required to evaporate the bound water. The heat loss of the overall drying system was calculated by multiplying total surface area of the oven with the heat loss coefficient and duration of the drying determined from the experimentally.

2.3. Grinding

One kg (on a wet basis) of both the raw and torrefied wood chips was ground using a laboratory heavy-duty hammer mill

(10HMBL, Glen mills). A computer with a data logger assembled with a multi-function transducer was connected to the mill to record the instantaneous power consumption during the grinding experiment. The samples were fed into the hammer mill using a vibrating feeder specifically designed for feeding material to hammer mill and pelletization equipment. The flow rate of the material was controlled to avoid exceeding the maximum power of the machine. A circuit breaker was installed in the grinder motor to avoid any overloading. Four trials were run for both torrefied wood chips and raw wood chips and the average time required for completing the grinding of 1 kg of the material was determined to calculate the flow rate. The 1/16″ mesh was used to screen the particles. During grinding, the instantaneous power consumption data with respect to time was recorded and used for calculating the required energy. The energy consumption (J/g) was calculated by dividing the power consumption (J/s) by the flow rate (g/s). We have also run the mill for 3 min without feeding any material to record the power consumption of the blank run [32,33].

2.4. Torrefaction

Torrefaction was carried out using the UBC-BTGA (University of British Columbia's big thermo-gravimetric analyser). The schematic representation of the set-up is presented in Fig. 1. The unit consists of a Carbolite oven (650 × 480 × 410 mm), which operates at temperatures ranging from room temperature to 600 °C. The heat convection inside the oven was facilitated by using a RPM adjustable fan. Torrefaction chamber (385 × 385 × 330 mm stainless steel box) was installed inside the oven and an oxygen-free environment for the chamber was provided by continuous flow of nitrogen. There are two ports for gas input and output. An external heater initially heats up the gas, then the preheated gas enters oven and flows through a heat exchanger. The preheated nitrogen gas was then purged onto the box. The gas exit port was connected to both the load cell and gas condensation unit with a tube. The gas carries the volatiles released from torrefying the sample through externally heated tubes, which were well insulated to prevent condensation and in-line deposition of the tar. The condensation unit was connected to the end of this tube which was cooled down using the cold water circulation outside the tube.

A tray (~350 mm diameter) is suspended freely inside the air sealed chamber via a thin wire from a load cell located about 1 m from the top of the chamber. The tray can generally hold up to 200–600 g of wood chips and we have used 300 g of wood chips/pellets to form a uniform layer on the tray with a height of approximately 1.8 mm and the load cell converts the force to signals and a data accusation card installed on the BTGA unit computer receives the data and mass loss was recorded instantaneously.

2.5. Pelletization

Lab scale California pellet mill (CPM) was used for pelletizing both torrefied and untreated materials (CL-397179, 230 V, 3 phase and 60 Hz). The pelletizer has 5 HP motor using 14 A and has a rotation speed of 1750 rpm. The ampere indicator was used to monitor the instantaneous power consumption of the machine. Similar to the grinder, the pelletizing machine was also connected to computer for recording the power consumption during the process. The flow rate of the material was controlled by adjusting the vibration of the feeding plate thus avoiding exceeding the maximum power consumption of the machine. The ground untreated material had 13% moisture content prior to pelletisation. Since the torrefaction process reduce the moisture content of the material to less than 1%, the material had to be re-conditioned to 13% moisture subsequent to torrefaction and grinding. In addition, the pelletisation of torrefied material was carried out with and without the use of binding agents. In the experiments using binders, the torrefied and subsequently ground sample was supplemented with 7 wt% binder (wheat flour) and thoroughly mixed for 20 min to ensure uniform distribution of the binding agent. The conditioned materials were fed to the pelletizer using vibrating feeder.

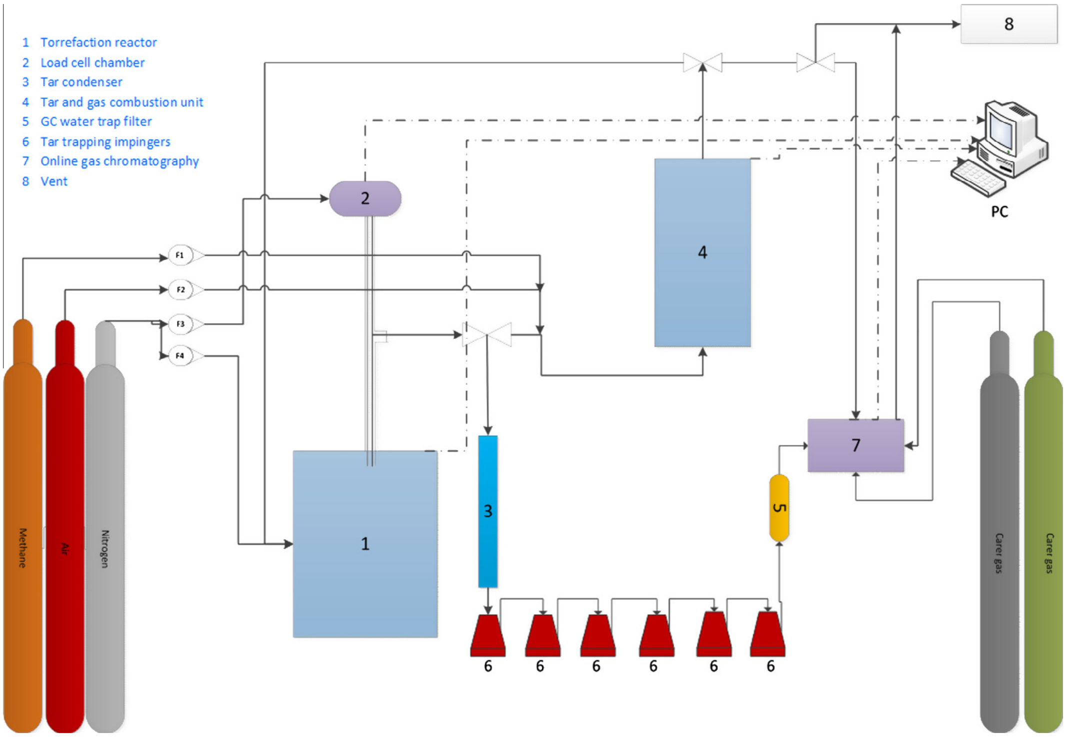

Fig. 1. Schematic representation of the thermogravimetric analyser set-up used in the work.

2.6. Determination of pellet properties

The pellet density was measured by dividing its mass over the pellet volume. The volume was calculated by measuring pellet diameter and length using a digital caliper [34]. Moisture content of the pellets was determined using the convection drying oven method according to the ASABE standards. The durability was measured using the tumbler testing unit according to American Society of Agricultural Engineers Standard S269.4 [35]. The unit consisted of a rectangular stainless steel container with inner dimension of 300 × 300 × 125 mm. One baffle inside the container enforced the tumbling effect and the rotation speed is adjusted to 50 rpm and the rotation time is 10 min. 500 g of sample was used for the test. The treated sample was subsequently sieved using round screen holes of 3.15 mm and the percent of original pellets that remained unbroken provided the durability index.

Moisture absorption rate of pellets was measured by placing them in a controlled humidity chamber (Model LHU-113, ESPEC Corp., Japan) [34]. The pellet samples were completely dried prior to analysis. The chamber temperature was set at 30 °C with a relative humidity (RH) of 90%. The increase in mass of the sample was monitored and recorded every 5 min in the first 30 min and every 1 h afterwards. The weight was recorded until it reached a constant value. The water uptake test was performed by immersing a given amount of pellets in water at 1:5 solid to liquid ratio for 30 min. The excess water was subsequently filtered out using 500 μm nylon mesh. The wet residue was subsequently weighed and completely dried at 105 °C till constant weight. The weight loss during drying was recorded every 5 min in the first 30 min and every 10 min afterwards.

The higher heating value (HHV) of solid samples was determined in an oxygen bomb calorimeter (Model 6600, Moline, IL) fitted with continuous temperature recording. A small mass 0.6–1.2 g of densified sample was placed in the instrument chamber. The measurements were repeated three times on three different pellets. Elemental analysis of the samples was done using Carlo Erba Elemental Analyzer (EA1108). The pellets were also analysed for their chemical constituents such as acid insoluble lignin using the Tappi-T-22 om-88 as previously described [36]. The hydrolysate from this analysis was retained and analysed for soluble carbohydrates [37] using Dionex (Sunnyvale, CA) HPLC (ICS-3000) equipped with an AS 50 auto sampler, ED50 electrochemical detector, GP 50 gradient pump and anion exchange column (Dionex CarboPac PA1).

3. Results and discussion

3.1. The two process configurations compared in the work

The two process schemes compared and evaluated in the work are outlined in Fig. 2. First we have determined the energy consumption in individual process steps of both the process schemes. Thereafter, we have compared the energy consumption and material balance when processing 1 kg of starting wood chips under the two different process scenarios. Subsequently, we have compared both the process routes for their ability to improve the pellet properties.

3.2. Energy consumption in individual process steps

3.2.1. Drying

In the process scheme I and II, wood chips were first dried to 15% using the hot air convection oven. The total energy required to reduce the moisture content from 40% to 15% was 1328 kJ/kg of the starting wet wood chips. Although this energy requirement was apparently similar for both process schemes, for drying below 15%, the process scheme II required lower amount of external energy compared to process scheme I. For the process scheme 1, drying step was immediately followed by the torrefaction step. Therefore, all of the remaining 15% moisture had to be removed to enable a direct torrefaction of the wood chips. This additional energy required to remove the final 15% moisture in the torrefaction unit was 320 kJ/kg of starting wood chips (on a wet basis). However, in the process scheme II, drying was immediately followed by grinding and pelletisation and these two processes are accompanied by a reduction in the moisture content from 15% to 7% prior to torrefaction. This means energy required to remove 8% of the original moisture is saved by following the process scheme II and the final torrefaction step had to only remove 7% moisture present in the pellets. Additional energy required to drive of this remaining 7% moisture in the torrefaction unit was apparently lower than process scheme I and was calculated to be 144 kJ/kg of the starting wood chips on a wet basis. Therefore, the combined total energy for drying the material for the process scheme I and II were 1647.3 and 1471.4 kJ/kg of the starting wood chips.

3.2.2. Grinding

Grinding step exhibited considerable differences in energy consumption between the two process schemes. In the process scheme I, grinding was carried out on the torrefied wood chips whereas in process scheme II, the raw wood chips with 15% moisture content were subjected to grinding. Since biomass is generally flexible and tenacious, it is generally energy intensive to reduce the particle size prior to the pelletization process or using them in pulverized combustion systems [38]. Apparently, this high energy requirement was substantially decreased after the torrefaction of wood chips (Fig. 3). Our results confirmed many of the previous work, which reported that loss of moisture and some of the volatiles during torrefaction results in the loss of visco-plastic properties making biomass more brittle and easy to grind [19,21,22,38,39]. In this work, the total energy required to grind 1 kg of torrefied wood chips was found to be 39 kJ/kg compared to 292 kJ/kg required for grinding the untreated wood chips (Fig. 3). The values are close to what has been reported earlier by Bergman [40] whose work showed that torrefaction can reduce 70–90% energy requirements for size reduction.

3.2.3. Pelletisation

In the process scheme I, pelletisation was carried out on torrefied and subsequently ground wood chips whereas in process scheme II, pelletisation was carried out on the raw and ground wood chips. The torrefied wood chips had only 3% moisture after grinding and therefore had to be conditioned with water to reach a final moisture content of 12% prior to pelletisation. However, in the process scheme II, since the ground biomass already had a moisture content of 12%, moisture conditioning step was not required.

Torrefaction had a significant influence on the ease of pelletisation (Fig. 4). While it was possible to densify the untreated Douglas-fir with reasonable energy consumption, it was extremely difficult to densify the torrefied material albeit the requirement of higher energy consumption. The energy required to pelletize 1 kg of the untreated and torrefied material (on a wet basis) was 757 kJ/kg and 1164 kJ/kg respectively. Despite this 1.5 times higher energy consumption, ~15% of the torrefied material still came out of the machine non-pelletised whereas almost all of the untreated material (>98%) could be pelletised. The results indicated that binding characteristics of biomass was significantly lowered after it has been torrefied. Stelte et al. [30] has reported that interparticle bonding was significantly lowered after the torrefaction

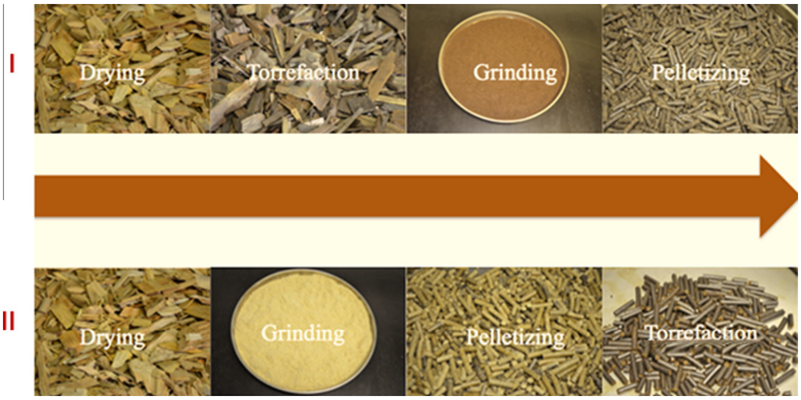

Fig. 2. Two different process configurations compared in the current work (process scheme I involve torrefaction prior to pelletisation and process scheme II involve torrefaction subsequent to pelletisation).

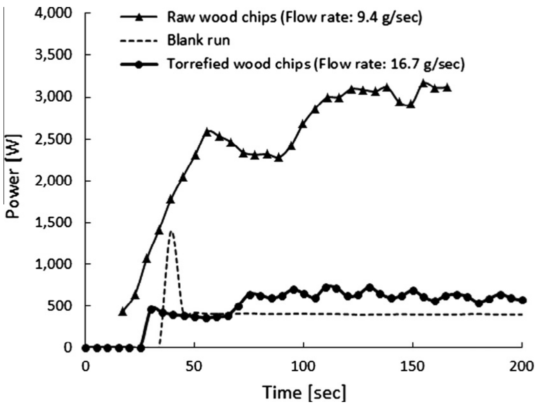

Fig. 3. Influence of torrefaction on the grinding energy requirements of Douglas-fir wood chips.

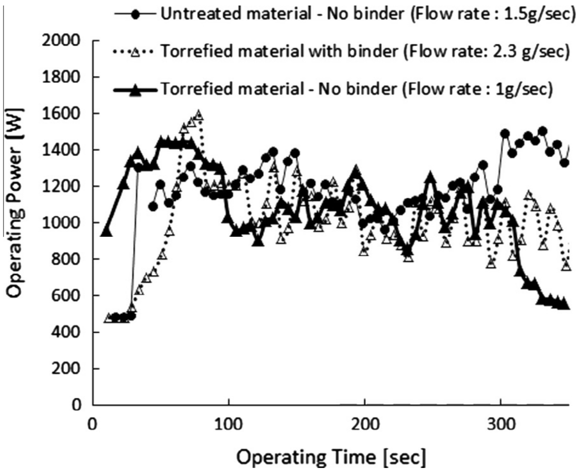

Fig. 4. Influence of torrefaction on the flow rate and power consumption during pelletisation of ground wood chips.

of Spruce. The authors have also reported that torrefied particles can increase the friction in the press channel of the pellet mill.

As it was apparent that the torrefied material could not be effectively densified even with higher energy requirement, in the next step, we have investigated the influence of adding a binding agent to enhance the binding affinity of the torrefied material. Since starch has been reported as an effective binder for the densification of biomass particles [33], wheat flour was mixed thoroughly with the torrefied particles at a proportion of 7% (on a dry weight basis) and subsequently pelletised. As was expected, use of binder substantially enhanced the ease of pelletisation lowered the energy requirements (Fig. 4). Greater than 98% of the torrefied material could be pelletised and total energy requirement for pelletizing the torrefied material was lowered from 1164 kJ/kg to 461 kJ/kg (Fig. 4). The results indicated that use of binding agent may be a prerequisite to enable the pelletisation of torrefied material with reasonable energy consumption. Previous work has also indicated the high energy requirement for densifying the torrefied biomass. Peng et al. [20,28] has shown that a high die temperature of 170–230 °C was required to enable effective densification of torrefied particles. [30] in their work on pelletisation properties of torrefied Spruce and Gilbert et al. [41] in their work on the pelletisation of herbaceous crops have also reported similar challenges with densifying the torrefied biomass after it has been torrefied [29,30,41].

3.2.4. Torrefaction

Torrefaction in the process scheme I was carried out directly on wood chips while in the process scheme II, the wood pellets were torrefied. The torrefaction was carried out under similar process conditions of 260 °C for 15 min. Prior to doing torrefaction, we had expected significant differences between wood chips and wood pellets for their response to the same set of torrefaction conditions. Since there are considerable variations between these two materials in terms of their density, surface area, bed voidage, etc. we had anticipated some differences in heat and mass transfer during the torrefaction of wood chips vs. wood pellets. Interestingly, the TGA results indicated that dry matter loss was fairly similar between the two feedstocks resulting in 14.9% and 15.1% dry weight loss respectively (Fig. 5). Apparently, the energy consumption to torrefy 1 kg dry weight of the wood chips and pellets were also very similar between the two process schemes and was 545 and 523 kJ/kg dry weight of the wood chips and wood pellets respectively.

3.3. Net energy consumption and mass balance

Fig. 6 summarises the sequential energy requirements for each step when starting with 1 kg of Douglas-fir wood chips (on a wet basis) via process schemes I and II. Process schemes I and II had a net energy input of 2.7 and 2.5 MJ/kg of wet wood chips processed (Fig. 6). While the energy required for torrefaction step was similar between the process schemes, energy requirement for drying and pelletisation steps was higher for process scheme I (Fig. 6). The higher energy requirement for drying in process

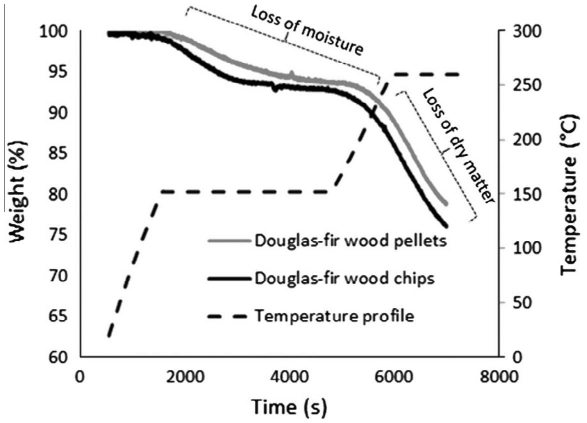

Fig. 5. Thermo gravimetric profiles of Douglas-fir wood chips and wood pellets during torrefaction.

scheme I was primarily because of the necessity for complete removal of moisture prior to torrefaction, whereas in process scheme II, partial reduction in the moisture content (~7%) was accomplished during grinding and pelletisation prior to reaching the torrefaction step. Similarly, the energy required to pelletize the torrefied material was also significantly higher than that of raw wood chips. Binding affinity of the raw biomass particles seems to have been substantially reduced after torrefaction [30]. Although the higher energy requirement in process scheme I was partly compensated by reduction in the energy consumption for grinding the torrefied wood chips, overall net energy consumption was still higher for process scheme I. Use of binding agent during pelletisation of torrefied material, however had significantly decreased the energy requirement for process scheme I from 2.7 to 2.3 MJ/kg, even less than that of the process scheme II (Fig. 6).

Although the present work holds great significance in comparing the two process pathways, the absolute energy values obtained in this work would vary significantly in an industrial system depending on the scale of operation, type of the equipments and reactors, their capacity utilisation, different energy profiles of each country/location. This may be a limitation of the current work. Although grinding and pelletisation steps consume electrical energy, drying and torrefaction can utilise different forms of energy. For example, both biomass and natural gas have been used as energy sources for drying/torrefaction. In addition, heat can be generated by the combusting the volatiles (gas and tar) released during torrefaction process and this heat can be integrated back to make the system auto thermal [40]. Such a comprehensive energy analysis would be highly relevant in a later stage to carry out technoeconomic assessment of an industrial scale operation and optimize the commercial process scheme. Location of the plant would also play a significant role in such an analysis since the energy resources and government policies would vary significantly with each region and thus the form of energy used and the mode of energy integration. However, it is important to mention that these factors should not outweigh the advantage of pathway II over pathway I presented in the current work since the main difference in the energy consumption between the two pathways arises from grinding and pelletisation steps which consumes same form of energy (ie, electrical energy).

The material balance for processing 1 kg of the wet wood chips (0.6 kg dry weight of the wood chips) via process schemes I and II are summarised in Fig. 6. The process scheme I resulted in 0.45 kg of torrefied wood pellets with 8% moisture content, which corresponded to 65% mass recovery on a dry weight basis of the starting wood chips (Fig. 6). The highest mass loss (15%) was occurred during pelletisation and this fraction of the torrefied material came out of the machine non-pelletised. The remaining mass loss was mainly from the torrefaction step (~15%). Use of binder during the process scheme I had increased the overall mass recovery to 85.3% (on a dry weight basis) of the starting material, however 6% of which was binder, therefore, the recovery of the original dry matter was 79% (Fig. 6). The increase in the mass recovery was mainly because; the use of binder had enabled the pelletisation of more than 98% of the torrefied material (Fig. 6).

The process scheme II resulted in a mass recovery of 81% of the dry weight of the starting material (Fig. 6). The loss of dry weight was mostly occurred during the torrefaction step. In addition to the difference in the loss of dry weight, one major difference between process schemes I and II was the difference in the moisture content between the final torrefied pellets. Since the torrefied material had to be conditioned for 12% moisture content prior to pelletisation, the process scheme I resulted in torrefied pellets with a moisture content of ~8%. However, in process scheme II, the raw wood pellets with 8% moisture was torrefied as the final step. Therefore, the resulting torrefied pellets were moisture free, however upon equilibrium with atmospheric humidity, the pellets had absorbed 3% moisture by weight. This difference in moisture implied that pellets generated by process scheme I will likely have an additional burden of transporting excess moisture and poor storage stability.

In addition to the energy input and mass balance, another important aspect which would help us choose a better process scheme is the quality of the resulting pellets. Therefore, we next compared both the process routes for their ability to improve the pellet properties that are critical to the transportability and stability of the pellets. As the process scheme I without the use of binder resulted in poor material recovery and higher energy consumption, we have chosen the process scheme I with the use of binders for comparing the pellet qualities to that of process scheme II.

3.4. Comparison of the pellet qualities

The pellets made from torrefied materials were denser (1207 kg/m^3) compared to the torrefied pellets produced by process scheme II (Table 1). In the process scheme II, the density of the wood pellets was decreased from 1158 kg/m^3 to 1031 kg/m^3 after torrefaction (Table 1). As the torrefaction was the final step, the process can increase the porosity of the raw wood pellets due to the volatilisation of the hemicellulosic components from the lignocellulosic matrix. However, despite a decrease in the density, the torrefaction did not change the shape or integrity of the wood pellets. In addition, as was mentioned earlier, the moisture content of the "pellets which was subsequently torrefied" was only 3% compared to 8% of the moisture present in the "torrefied and subsequently pelletised" material (Table 1). The durability of the torrified pellets was also fairly close and comparable to that of the raw material pellets (Table 1).

In order to understand the stability of the pellets in water, both types of pellets were subsequently immersed in water (Fig. 7a1 and b1). Compared to the raw material pellets, which readily disintegrated when immersed in water, both types of torrefied material exhibited substantially higher hydrophobicity. Among the two different types of torrefied pellets, the "torrefied and subsequently pelletised material" were relatively easily fall apart whereas the "pelletised and subsequently torrefied material" stayed intact for more than 2 h in water (Fig. 7a2 and b2). Given the results obtained in this work, there may be a possibility to enhance the hydrophobicity of the pelletised and subsequently torrefied material by doing further optimisations on a pilot scale.

In addition to the visual appearance, the hydrophobicity of the two torrefied pellets were further verified by drying the water immersed pellets and determined how much water was originally

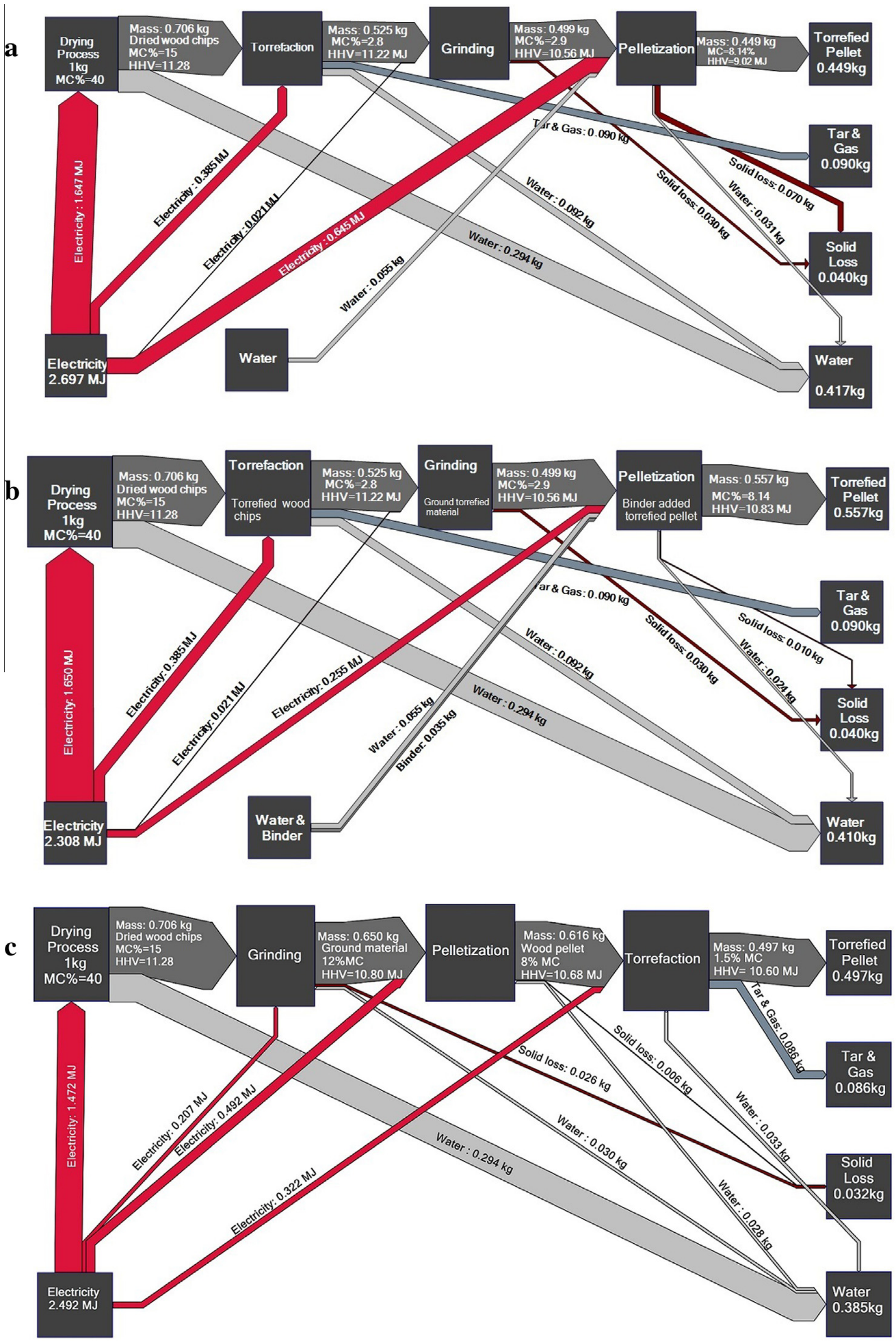

Fig. 6. Influence of the different process pathways on the material balance and energy consumption of making torrefied pellets. (a) Torrefaction prior to pelletisation without any binder. (b) Torrefaction prior to pelletisation with the use of binder. (c) Torrefaction subsequent to pelletisation.

Table 1
Influence of carrying out torrefaction before or after pelletisation on the durability of torrefied wood pellets.

Sample	Density (single pellet) (kg/m^3)	Moisture content (%)	Durability[a]
Raw material pelletised	1158.5	8.2	98.2
Raw material, pelletised and subsequently torrefied	1031.2	2.9	97.0
Raw material, torrefied and subsequently pelletised[b]	1207.3	7.7	98.6

[a] Measured by the tumbler method.

[b] Binder was used for the pelletisation of torrefied material.

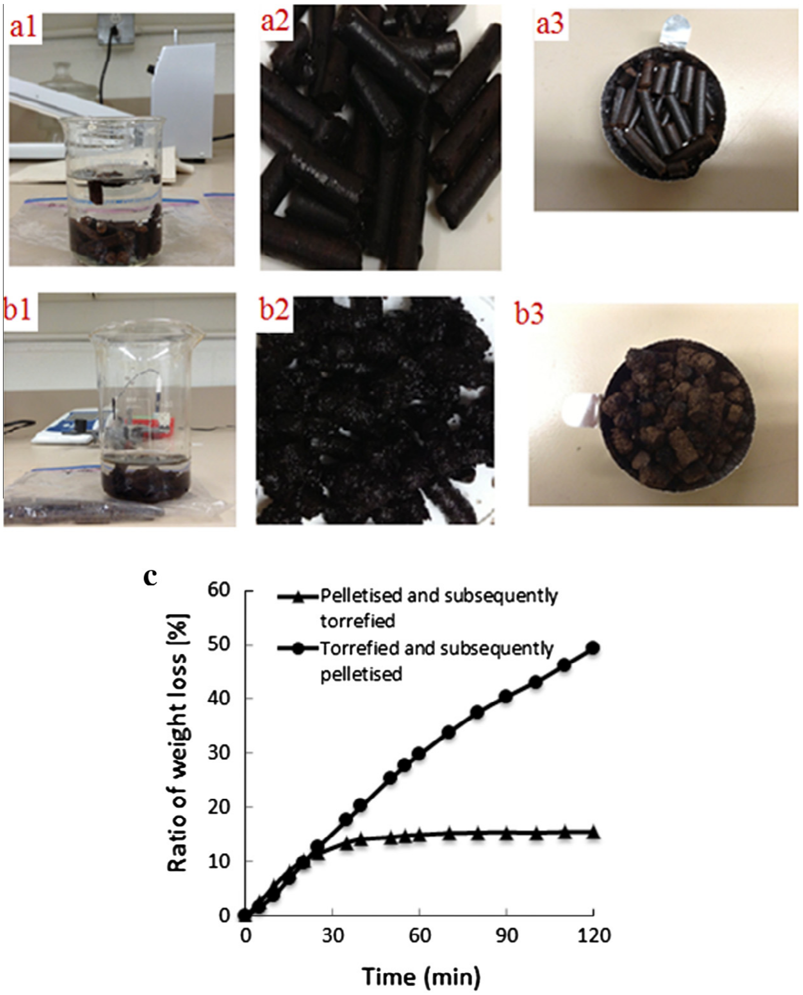

Fig. 7. Influence of carrying out torrefaction before or after pelletisation on the water absorption of the resulting pellets (upon immersion in water). (a) Pelletised and subsequently torrified material when (a1) immersed in water (a2) when taken out of water after 6 h (a3) oven dried after taking out from water. (b) Torrefied and subsequently pelletised (b1) immersed in water (b2) when taken out of water after 6 h (b3) oven dried after taking out from water. (c) Weight loss of the water immersed pellets (a and b) during drying.

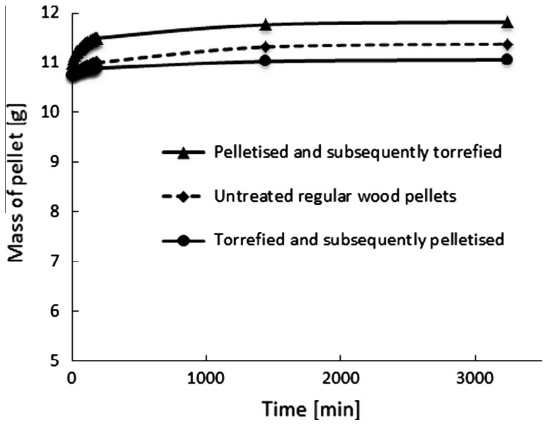

Fig. 8. Influence of the order of torrefaction and pelletisation on the moisture absorption capacity of the torrefied pellets.

Table 2
Influence of torrefaction in enhancing the carbon content (% dry weight basis) and heating value of Douglas-fir wood chips and wood pellets.

	Carbon	Hydrogen	High heating value (MJ/kg)
Douglas-fir wood chips	45.7	6.4	18.7
Douglas-fir wood pellets	46.5	6.4	18.7
Pelletised and subsequently torrefied	51.2	6.0	22.0
Torrefied and subsequently pelletised	49.5	5.4	20.8

absorbed by the pellets and how easily the absorbed water can be evaporated. We have observed that the torrefied and subsequently pelletised material absorbed substantially higher amount of water compared to the pelletised and subsequently torrefied material (Fig. 7a3 and b3). It appears that the higher hydrophilicity of starch being a binder can lead to an easy loosening of the pellets when immersed in water. This initial loosening may provide more access to water molecules to the interior regions of the pellets. In addition, it should be noted that wood chips during torrefaction (particularly at milder conditions) can lead to an uneven torrefaction with exterior of the wood chips being more hydrophobic and interior being more hydrophilic. These torrefied wood chips when subsequently ground will destroy the hydrophobic outer layer and expose new surfaces which may not be necessarily as hydrophobic as the outer layer of the torrefied wood chips. Therefore, one can expect a higher degree of hydrogen bonding between water and the torrefied particles in the case of ‘‘torrefied and subsequently

Table 3
Influence of torrefaction on the chemical composition (carbohydrates and lignin content) of Douglas-fir wood chips and wood pellets.

	Acid insoluble lignin/residue[a]	Arabinan	Galactan	Glucan	Xylan	Mannan
Douglas-fir wood chips	31.7 (0.4)[b]	1.1 (0.0)	3.3 (0.1)	43.2 (0.8)	5.0 (0.2)	12.1 (0.1)
Douglas-fir wood pellets	32.3 (0.7)	1.3 (0.0)	3.5 (0.0)	43.8 (0.6)	4.6 (0.0)	11.7 (0.3)
Pelletised and subsequently torrefied	43.3 (0.2)	ND[c]	0.7 (0.1)	49.1 (1.0)	0.9 (0.0)	4.6 (0.1)
Torrefied and subsequently pelletised	38.3 (0.5)	ND	0.5 (0.1)	54.7 (1.4)	0.8 (0.0)	3.9 (0.1)

[a] After torrefaction, the residue insoluble in 72% sulphuric acid may not be necessarily lignin.
[b] Deviations from the average of triplicate analysis.
[c] Not detected in the analysis.

pelletised" material. In contrary, in the material which is first "pelletised and then torrefied", the outer layer of the torrefied pellets is not destroyed and can remain hydrophobic preventing water to get access to the interior of the pellets.

We have also assessed the stability of the wood pellets in high humid conditions by exposing the pellets in the humidity chamber. The results showed that the degree of moisture uptake was in the order "pelletised and subsequently torrefied material" (process scheme II) > untreated regular pellets > "torrefied and subsequently pelletised" material (process scheme I) (Fig. 8). It appears that penetration of the water vapour and its subsequent condensation is critically dependent on the external surface area of the pellets. In the case of pelletised and subsequently torrefied material, it appears that torrefaction being the second stage substantially increased the external porosity of the pellets and more water vapour can potentially get condensed in the small pores present in the outer surface of the pellets albeit having a hydrophobic surface. In the case of untreated pellets and "torrefied and subsequently pelletised material", pelletisation being the last step resulted in the closer packing of the materials with less external surface area and porosity available for the water vapours to condense (Fig. 8).

Since the suitability of the torrefied wood pellets in subsequent applications will be largely governed by the chemistry of biomass, we have carried out a preliminary elemental analysis and chemical composition of the samples (Tables 2 and 3). As was expected, the elemental analysis results showed an increase in the percentage of carbon and therefore an apparent increase in C/O ratio after torrefaction (Table 2). The increased carbon to oxygen ratio was also in agreement with the increased calorific value of the torrefied biomass compared to the raw material (Table 2). Despite a decrease in the hemicellulose content as shown by the chemical compositional analysis (Table 3), the carbon content seems to have mostly preserved during torrefaction. The results indicate that the most of the carbon present in the hemicellulose was transformed and detected as the acid insoluble residue during the compositional analysis. The 'torrefied and subsequently pelletised' (pathway I) material has shown a reduction in the carbon content probably due to the addition of binder. Starch has relatively lower C/O ratio compared to lignin and therefore supplementation of wheat flour can reduce the proportion of carbon in the resulting pellets (Table 2). The mass percentage of hydrogen was also slightly decreased after torrefaction. This decrease in the hydrogen with simultaneous increase in the carbon content can be considered as an indication of the increased hydrophobicity of the material as was confirmed by the earlier experiments. When considering the molar basis, the change in hydrogen content can be significant and would be influential enough to reduce the hydrogen bonding cites. With the addition of the binder, the amount of hydrogen seems to have lowered again, however due to the increased proportion of oxygen present in starch.

The data on chemical composition showed that pelletisation alone did not substantially change the chemical composition of the raw material. The total carbohydrate content of raw wood chips and raw wood pellets were close to 65% (Table 3) which is typical of most of the softwood species [38]. However, as was expected, torrefaction substantially reduced the hemicellulose content of the raw wood chips as well as the raw wood pellets indicating their volatilisation and carbonisation during the non-oxidative torrefaction process [42,43]. It should be noted that loss of hemicellulose can reduce the hydrogen bonding sites in the material leading to a lower affinity to water. This also explains why torrefied material was more hydrophobic compared that of the raw material. The relative increase in the percent lignin content of the material is apparently due to the volatilisation of most of the hemicellulose components and formation of the degradation products which are insoluble in acid [44,43]. Therefore, the properties of this insoluble residue might be substantially different compared to the properties of lignin present in the raw material [44]. The higher glucan content of the "torrefied and subsequently pelletised" samples could be from the added starch as the binder. Since significant fraction of the carbohydrates were still preserved in the material at these torrefaction conditions, it would be interesting to see the suitability of these feedstocks for biochemical conversion applications.

4. Conclusions

The described work compared two different process routes to convert Douglas-fir wood chips to torrefied wood pellets as the final product. It was possible to successfully combine torrefaction and densification processes to improve both energy density and bulk density of biomass thus producing "renewable biocoal" that can be used as a tradable commodity to potentially substitute fossil fuels. Although traditionally the wood chips are first torrefied and subsequently densified, this particular work showed that it was more energy efficient to first densify the wood chips and subsequently torrefy them. However, the use of binder in the conventional process scheme can significantly reduce the energy consumption while resulting in pellets with enhanced breaking strength, durability and density. However, use of binder compromised some other qualities of the pellets with the pellets produced by process scheme II being still superior in terms of their high stability in water, enhanced calorific value, higher carbon content and reduced moisture content. Therefore, "Torrefaction subsequent to pelletisation" currently appears to be a promising strategy to obtain torrefied wood pellets which has reasonable transportable properties with high potential in different end product applications.

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